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Decreasing Trend in Formaldehyde Detected From 20-Year Record at Wollongong, Southeast Australia

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Abstract

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Decreasing Trend in Formaldehyde Detected From 20-Year Record at Wollongong, Southeast Australia

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Key Points:
- Significant decrease in HCHO from 1996 to 2015 is observed at Wollongong in all months but November, superimposed on a regional HCHO increase
- Decrease is linked to changes in summer biomass burning and declining local industrial emissions
- Lack of trend in November is linked to rising temperature and earlier onset of biogenic emissions

Supporting Information:
- Supporting Information S1

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Abstract The response of atmospheric composition to ongoing environmental change remains poorly constrained across much of the Southern Hemisphere. We use a 20-year record of ground-based total column measurements from Wollongong, southeast Australia to identify a statistically significant decreasing trend in formaldehyde of −1.9 [−2.2, −1.7]%/year. The trend is consistently negative across all months except November. Satellite data indicate that the trend at Wollongong is distinctly local and is superimposed on a regional-scale increase likely driven by changes in methane. In austral summer, coincident changes in hydrogen cyanide suggest that decreases in local biomass burning can only partly explain the observed trend. In the absence of other explanations, we infer that the observed formaldehyde trend is likely driven by decreasing industrial emissions. In November, an observed increasing temperature trend is consistent with an earlier onset of biogenic emissions in the region, driving increased biogenic formaldehyde that counteracts the overall decrease.

1. Introduction

Formaldehyde (HCHO) is a short-lived tropospheric pollutant and high-yield oxidation product of non-methane volatile organic compounds (NMVOCs; Franco et al., 2016; Millet et al., 2008; Pfister et al., 2008). Observed HCHO measured from ground-based and satellite remote sensing platforms is frequently used to infer the fluxes of NMVOCs, in particular, isoprene, and can provide insight into how these fluxes are responding to ongoing environmental change (Abbot et al., 2003; Barkley et al., 2009; Marais et al., 2012; Millet et al., 2008; Zhu et al., 2014). Southeast Australia is a global hot spot for both HCHO and isoprene (Guenther et al., 1995; Marais et al., 2012; Pfister et al., 2008; Stavrakou et al., 2009a) and is also experiencing rapid and extreme changes in climate (Hughes, 2003; Bindoff et al., 2013; Murphy & Timbal, 2008) that could have a significant impact on isoprene emission, HCHO production, and subsequent atmospheric chemistry. Here, we use a long-term record of HCHO from a ground-based remote sensing instrument at Wollongong, New South Wales, Australia to quantify recent trends in HCHO and investigate their likely drivers.

On a global scale, the main source of atmospheric HCHO is the oxidation of methane (CH_4) by the hydroxyl radical ($^{\cdot}OH$; 45–60%), followed by the oxidation of NMVOCs (20–30%; Dufour et al., 2009; Franco et al., 2016; Pfister et al., 2008; Stavrakou et al., 2009b). Direct emissions from fuel combustion, biomass burning and vegetation account for only a minor part of the global HCHO budget but can be important local sources, particularly during fire events (Dufour et al., 2009; Fortems-Cheiney et al., 2012; Franco et al., 2016; Jones et al., 2009; Holzinger et al., 1999; Pfister et al., 2008; Stavrakou et al., 2009a, 2009b). HCHO has a short lifetime of only a few hours against photolysis and reaction with $^{\cdot}OH$ (Fortems-Cheiney et al., 2012; Zeng et al., 2015). This makes it an effective indicator of local sources, both primary and secondary (Jones et al., 2009; Marais et al., 2012; Palmer et al., 2001). Over Australia, models and satellite data suggest that 40–80% of HCHO is produced from the oxidation of biogenic NMVOCs, with maximum contributions in the southeast part of the country (New South Wales) where eucalypt-dominated ecosystems are prevalent (Dufour et al., 2009; Franco et al., 2016; Pfister et al., 2008). However, these regional estimates have not been verified with observations of HCHO.

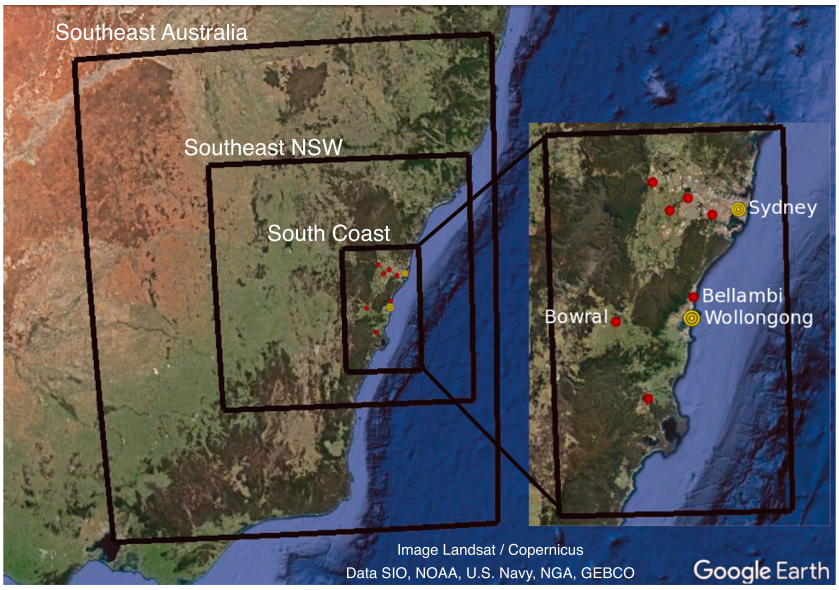

Figure 1. Map of southeast Australia showing the location of the Fourier Transform Infrared Spectrometer at Wollongong, Bureau of Meteorology sites (red) and three regions over which satellite data were averaged (black boxes). The city of Sydney is also shown for context.

Recent changes in temperature, global CH_4 abundance, and possibly emissions of HCHO and its precursors are likely driving changes in HCHO abundance, but the magnitude and effects of these changes in southeast Australia are unknown. Between 1910 and 2010 temperatures in Australia increased 0.8 °C (Pink, 2012), and both biogenic emissions and oxidative chemistry would be sensitive to this change. Globally, methane abundance increased over the years 1984–1999, stayed almost steady from 1999–2006, and then resumed growing from 2007 onwards (Nisbet et al., 2014) with implications for HCHO production (Franco et al., 2016). Decreases in biomass burning emissions have been suggested to explain trends in carbon monoxide over Australia from 2002–2011 (Yin et al., 2015), and declining industrial emissions from 1997–2009 have been shown to affect both carbon monoxide and ethane at nearby sites in New Zealand and Antarctica (Zeng et al., 2012); both of these emission trends could also impact HCHO in southeast Australia.

A long-term record of HCHO abundance available from a ground-based Fourier Transform Infrared Spectrometer (FTIR) measuring solar absorption spectra at Wollongong, New South Wales (Figure 1) can provide insight into the implications of these environmental trends. While short periods of this data set have been used to evaluate models (Zeng et al., 2015), the full 20-year data set has not previously been leveraged for understanding atmospheric change in a biogenic-dominated region. This record provides a unique platform to probe changes in a global biogenic hot spot.

Here we evaluate variability and trends in HCHO in southeast Australia, using Wollongong FTIR measurements collected between May 1996 and December 2015. Section 2 describes the measurements and identifies a statistically significant decreasing trend over the 20-year observational period. In section 3, we use a suite of ground-based and satellite observations of HCHO and related parameters to investigate the drivers of the decreasing HCHO trend. Finally, conclusions are presented in section 4.

2. HCHO Observations and Trend Detection

HCHO total column measurements were collected with the FTIR at the Wollongong site of the Network for the Detection of Atmospheric Composition Change (NDACC), located at the University of Wollongong (34.45° S, 150.88° E, 30 m above sea level, see Figure 1). Measurements are derived from ground-based solar infrared absorption spectra (Griffith et al., 1998; Té et al., 2016) following the procedure outlined in Jones et al. (2009) as detailed in section S1 in the supporting information. Figure 2a shows the individual retrievals of HCHO total column abundance from May 1996 to December 2015. The influence of previously identified

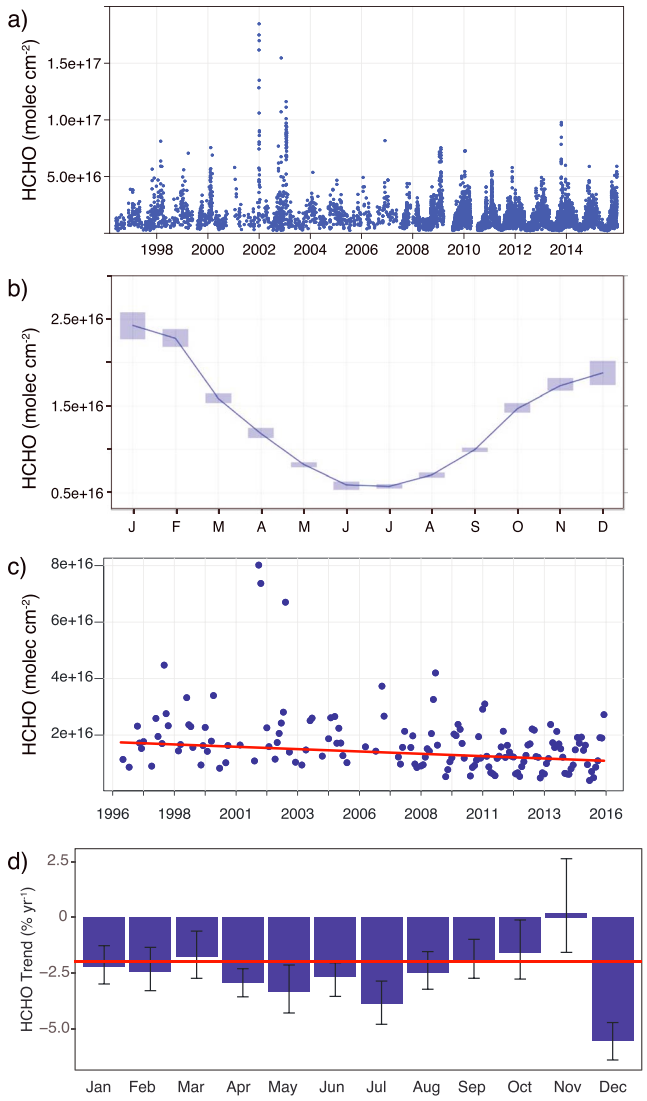

Figure 2. HCHO observations from Fourier Transform Infrared Spectrometer at Wollongong, southeast Australia from May 1996 to December 2015 showing (a) the long-term time series, (b) the mean seasonal cycle, with shaded regions representing the 95% confidence interval of the observations, (c) the deseasonalized monthly mean anomalies (blue dots) and trend (red line), and (d) monthly trends and 95% confidence intervals computed as described in the text, along with the overall trend from (c) shown as the red line.

local fire events can be seen particularly in December 2001 to January 2002, January 2003, and October 2013 (Paton-Walsh et al., 2004, 2005, 2010; Rea et al., 2016). The mean seasonal cycle, shown in Figure 2b, shows that HCHO peaks in austral summer due to the large influence from biogenic NMVOC emissions at this time of year (Jones et al., 2009; Zeng et al., 2012).

Trends in HCHO abundance were calculated using the TheilSen function in the R package OpenAir (Carslaw & Ropkins, 2012). The Theil-Sen method is relatively insensitive to outliers and does not require residuals to be normally distributed (Gilbert, 1987; Helsel & Hirsch, 1992). Trends were calculated from deseasonalized monthly mean midday (11:00–15:59) total HCHO columns for months when at least five valid measurements were available (comprising 70% of all months with at least one valid measurement). As detailed in section S2 in the supporting information, we restrict the analysis to midday values and limit each monthly mean to five data points to account for a change in sampling rate caused by automation of the measurements in 2007. We used a bootstrapping procedure to repeatedly calculate the trend after recalculating the monthly means from five randomly sampled measurements in each month with sufficient data. Further details and statistical tests are provided in the supporting information. All reported trends are significant to $p < 0.05$ and are reported as the median of 10,000 (overall) or 1,000 (monthly) bootstrap iterations, along with the 95% confidence interval in brackets (derived from the bootstrap distributions).

Figure 2c shows the deseasonalized monthly mean midday (11:00–15:59) total HCHO columns for months with at least five valid measurements, along with the calculated HCHO trend of −1.9 [−2.2, −1.7] %/year. Using the monthly medians rather than monthly means resulted in a calculated trend of −1.8 [−2.1, −1.4] %/year. The magnitude of the trend is not particularly sensitive to our requirement of five valid measurements; changing the cutoff to four valid measurements (78% of all months with at least one valid measurement) resulted in a calculated trend of −1.8 [−2.0, −1.5] %/year and including all months with at least one valid measurement (no bootstrapping) resulted in a calculated trend of −1.7 [−2.1, −1.4] %/year. In all cases, the trend is consistently negative and statistically significant. To exclude the effects of the known large fire events noted above, we also calculated the trend from February 2003 to September 2013 and found that it increased in magnitude to −3.0 [−3.5, −2.4]%/year.

To elucidate possible causes of the observed HCHO trend, we calculated the trend in HCHO abundance for each month, as different source and sink influences have different seasonality. HCHO production from biogenic NMVOC sources peaks in austral summer (November–February; Zeng et al., 2015), while local biomass burning peaks in October–January (Edwards et al., 2006; Zeng et al., 2015), and anthropogenic emissions have limited seasonality (Helmig et al., 2009). Methane oxidation is largest in summer due to increased $^{\bullet}$OH production (Khalil & Rasmussen, 1983), but the impact on HCHO may be offset by more rapid HCHO oxidative loss. While the mean seasonal variation of HCHO at Wollongong (Figure 2b) is driven by the seasonality of biogenic NMVOC emissions, other influences not visible in the multiyear mean could impart a seasonal signature to the trends. Figure 2d shows that the monthly trends are strongly negative (between −1.6 and −5.5%/year) except in November, when there is no significant trend. With the exception of November, the monthly trends are largely consistent with one another and with the overall trend (shown in Figure 2d as the red line).

3. Diagnosis of Observed Trends

3.1. Methane Oxidation

On a global scale, CH_4 oxidation is the largest source of HCHO (~50%; Pfister et al., 2008). Changes in background CH_4 abundance have been shown to affect HCHO observed at other sites and could be impacting the HCHO observations at Wollongong. At Jungfraujoch, for example, Franco et al. (2016) found 1998–2015 changes in HCHO and CH_4 were strongly correlated. Using FTIR measurements, the authors found a decrease in HCHO abundance of −3.68 ± 1.00%/year from 1996–2002, the period when global CH_4 levels stabilized (Aydin et al., 2011; Dlugokencky et al., 2003; Simpson et al., 2012), and an increase in HCHO abundance of +0.81 ± 0.62%/year from 2003–2015, when CH_4 levels were stable followed by resumed growth (Kirschke et al., 2013; Nisbet et al., 2014).

The trends observed at Wollongong do not seem to be similarly affected by background changes in CH_4 oxidation. While the FTIR at Jungfraujoch samples the free troposphere where CH_4 oxidation is the dominant HCHO source, the Wollongong FTIR is just above sea level and is therefore more sensitive to local surface processes. In addition, the timing of the observed HCHO trends is not consistent with the timing of changes in global CH_4 abundance. From 2003–2015, when methane levels were stable then increasing (Nisbet et al., 2014) and HCHO increased at Jungfraujoch (Franco et al., 2016), Wollongong HCHO observations showed a large decrease of −2.6 [−3.0, −2.2] %/year.

We expect that any CH_4-driven change in HCHO would be at least regional in scale. To further explore the impact of CH_4 oxidation on changing HCHO in southeast Australia, we used satellite observations of HCHO total column measurements from the Ozone Monitoring Instrument (OMI) level 2 global swath product, OMHCHO (Gonzalez Abad et al., 2015; Zhu et al., 2016). For details of the instrument and retrieval, see section S1 in the supporting information. We chose OMI rather than other satellite products as it had the longest continuous overlap with the FTIR record. Satellite measurements were averaged daily and gridded to obtain mean HCHO abundance from 2005–2015 over the three regions shown in Figure 1. The "South Coast" region (33.5–35.5°S, 150.0–151.5°E) contains land within 100 km of Wollongong and limited ocean cover and was selected to represent the region over which isoprene and/or HCHO could be expected to travel within their lifetimes. The "Southeast NSW" (32–36°S, 147.5–152.5°E) and "Southeast Australia" (30–38°S, 145–153°E) regions cover the wider regional area. Due to the relative sparsity of both population and vegetation in southeast Australia (see Figure 1), HCHO columns are generally low across much of the region, and these regional averages are roughly indicative of the HCHO background (Figure S1).

The decreasing HCHO trend seen in the Wollongong FTIR data is not evident in the OMI data, suggesting it is a local rather than regional phenomenon. OMI measurements over the South Coast region showed no significant trend over the 2005–2015 period. The larger Southeast NSW and Southeast Australia regions showed increases in HCHO of +0.9 [+0.4, +1.6] %/year and +1.5 [+0.8, +2.0] %/year, respectively. These are opposite in sign to the trend calculated from the FTIR measurements which, when limited to the 2005–2015 OMI period, still showed a decrease of −2.6 [−3.2, −2.0] %/year in HCHO. Monthly trends were generally insignificant over this shorter period, except for an August increase in the OMI data for all three regions and January, May, and July–September decreases in the Wollongong FTIR measurements.

The observed regional-scale increase in OMI HCHO during this period is likely linked to changes in global CH_4 abundance, as seen elsewhere (Franco et al., 2016). The decreasing trend at Wollongong, however, is inconsistent with this increasing trend—suggesting that the greater regional increase in HCHO is being overpowered by local changes at Wollongong that cannot be attributed to CH_4.

3.2. Biogenic Emissions

In southeast Australia, models suggest that oxidation of NMVOCs is the dominant source of HCHO (~80%; Pfister et al., 2008). Isoprene accounts for approximately half of the total NMVOC emissions globally (Guenther et al., 2012), including in Australia (Emmerson et al., 2016). Biogenic emissions of isoprene are strongly temperature-dependent, with a threshold of around 12 °C below which no isoprene is emitted (Oku et al., 2014). Above this threshold, isoprene emissions increase substantially up to a thermal optimum of 35 to 40 °C (Monson et al., 1992; Sharkey & Yeh, 2001). HCHO abundance is therefore sensitive to temperature changes via emission of biogenic precursors.

We tested whether temperature in the Wollongong region showed decreasing trends consistent with the observed HCHO decrease using temperature observations from sites around Wollongong (see Figure 1)

provided by the Australian Bureau of Meteorology (BoM). Due to the short lifetime of both isoprene (typically less than 1 hr; Crutzen et al., 1999; Palmer et al., 2003) and HCHO (on the order of a few hours; De Smedt et al., 2015; Palmer et al., 2003), only sites within 100 km of Wollongong were considered close enough to be representative of biogenic emission regions that potentially influence the site. This distance should also be sufficient to account for slower HCHO production via low NO_x chemistry during transport of precursor NMVOCs.

We calculated overall and monthly trends for each BoM site using the daily minimum and maximum temperature data for all available dates from 1997–2015 (mean temperature data were not available). Taken together, the sites showed no clear indication of consistent temperature change over the region. Only a small fraction of the sites showed a trend in either the overall or monthly data, and these were not consistent across sites or across months. We also calculated the regional mean temperature trends using the observations from all sites with available data for 1997–2014 (including only days for which temperature data were available from all sites). While no overall trends were seen, in November there was an increase of +0.1 [0.0, +0.2] °C/year, equivalent to +0.7%/year.

We further investigated the possible role of temperature-driven changes in biogenic emissions using temperature observations from Bowral, an inland site located in a highly vegetated region approximately 45 km west of Wollongong (see Figure 1) that could be a large source region for biogenic HCHO precursors. Temperature measurements at Bowral were only available at 09:00 and 15:00 local standard time from January 1997 to January 2015. As with the multisite mean, no overall trend was detected. In November, however, the observations showed an increasing trend in temperature of +0.2 [0.0, +0.3] °C/year in the morning (09:00) and +0.2 [0.0, +0.4] °C/year in the afternoon (15:00), equivalent to +1%/year.

The lack of a consistent, decreasing temperature trend in the Wollongong region implies the observed HCHO trend is not caused by temperature-driven decreases in biogenic emissions. However, given the strong sensitivity of isoprene emissions to atmospheric temperature, it is likely that the November warming observed both at Bowral and in the multisite mean has driven an increase in November biogenic emissions in the southeast Australian region. Minimum temperatures in this area typically reach the threshold for isoprene emission (12 °C; Oku et al., 2014) sometime in November or December, and at Bowral the mean minimum temperature in November rose from below 12 °C to above 12 °C over this time period. The warming may be associated with both a higher magnitude and an earlier onset of isoprene emissions in the region in November, with subsequent oxidation leading to an increase in biogenically derived HCHO. We therefore suggest that the overall strong decreasing trend in HCHO observed during the rest of the year is being offset in November by an increase in biogenic HCHO - with the net effect being no significant change in November HCHO over the past 20 years.

Biogenic emissions are also sensitive to other environmental factors, including soil moisture stress, vegetation cover, and solar radiation. As no local measurements exist for these variables, we evaluated trends using satellite-derived estimates (Dorigo et al., 2017; Gruber et al., 2017; Liu et al., 2012; Yuan et al., 2011). As discussed in section S3 in the supporting information, none of these variables showed statistically significant trends that were consistent with the observed HCHO trends.

3.3. Biomass Burning Emissions

Biomass burning and anthropogenic emissions provide minor additional sources of HCHO. Primary emissions of HCHO from these sources account for only 0.4–0.8% of the global budget (Fortems-Cheiney et al., 2012; Pfister et al., 2008; Zeng et al., 2015), and secondary production from NMVOC emissions from these sources accounts for roughly 10% of the global budget (Shim et al., 2005; Stavrakou et al., 2009a). However, these sources can be important on local scales, particularly during large emission events such as major fires (Stavrakou et al., 2009a). To investigate the potential impact of biomass burning on HCHO at Wollongong, we compared the observed trends in HCHO to those for hydrogen cyanide (HCN) and carbon monoxide (CO), also measured by the Wollongong solar FTIR. While CO and HCN have longer lifetimes than HCHO, we expect any major changes in local biomass burning to be reflected in all three gases. We use HCN as a tracer of biomass burning, its primary source (Li et al., 2000, 2003, 2009). CO in this region is also sensitive to biomass burning but is additionally influenced by CH_4 and NMVOC oxidation and anthropogenic emissions (Fisher et al., 2015; Yin et al., 2015; Zeng et al., 2015). We use it here to provide an indication of whether changes in all three gases are similar, pointing to local biomass burning influence as a driving factor.

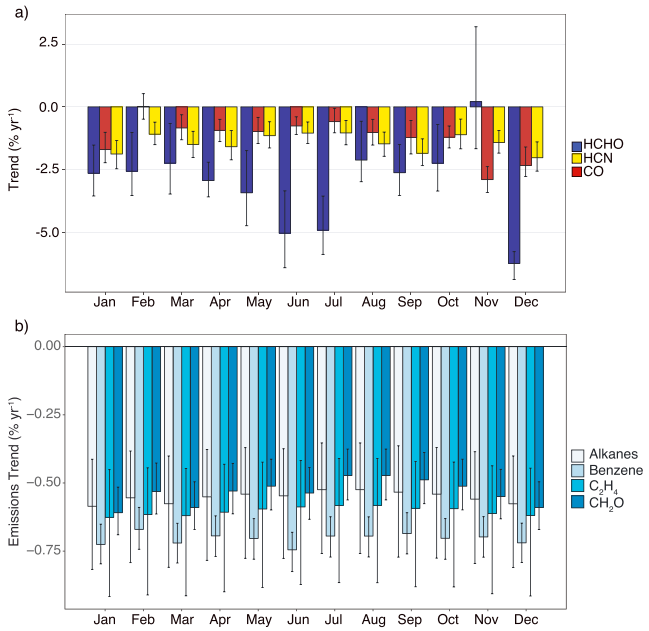

Figure 3. Monthly trends and 95% confidence intervals at Wollongong, Australia from 25 May 1997 to 2 April 2014 in (a) HCHO (blue), HCN (yellow), and CO (brown) total columns calculated from FTIR observations and (b) estimated anthropogenic emissions of HCHO and NMVOC precursors calculated from the CEDS emission inventory. FTIR = Fourier Transform Infrared Spectrometer; NMVOC = non-methane volatile organic compound; CEDS = Community Emissions Data System.

Figure 3a compares monthly trends in HCHO, CO, and HCN for the period of time over which measurements of all three gases overlapped (25 May 1997 to 2 April 2014). Over this period, the year-round abundance of all three gases decreased with trends of −2.4 [−2.7, −2.1] %/year for HCHO, −1.4 [−1.5, −1.2] %/year for CO, and −1.0 [−1.1, −0.84] %/year for HCN. Figure 3a shows that the monthly trends are generally negative for all three species, and HCN and CO trends are nearly always similar in magnitude. For both HCN and CO, large negative trends are seen in December–January, when local fires are frequent (Paton-Walsh et al., 2004, 2005, 2010; Rea et al., 2016; Zeng et al., 2015). This suggests that changes in biomass burning frequency and/or intensity are driving a decrease in both species that is strongest in austral summer. We further tested this hypothesis and in particular the role of local fire emissions using estimated biomass burning from the Global Fire Emissions Database version 4 (GFED4s) described in van der Werf et al. (2017), averaged over the South Coast region shown in Figure 1. The year-round GFED4s trend was negative but not statistically significant (−2.7 [−5.1, 0.0] %/year). On monthly timescales, statistically significant decreasing trends were found for January–June (−2.2 to −3.9%/year), September–October (−5.3%/year), and December (−8.5%/year).

Based on the HCN, CO, and GFED4s trends, we expect that some fraction of the observed HCHO decrease can be attributed to decreasing biomass burning in southeast Australia, particularly in December when all three species and the emissions show the largest decreasing trend. Biomass burning changes are not, however, consistent with all observed features of the decreasing HCHO trend. Outside of December, HCHO trends are strongest in austral winter, when HCN and CO trends are weak and GFED4s actually shows a small (but not statistically significant) increase in biomass burning. Further, we showed in section 2 that when we exclude the effects of known fire events by calculating the HCHO trend from February 2003 to September 2013, the decreasing trend becomes stronger by 50%, changing from −1.9 to −3.0%/year. Over this time period, GFED4s showed a weak, positive, and statistically insignificant trend in biomass burning (+0.68 [−3.0, +4.5]%/year). This suggests that the overall negative trend in HCHO at Wollongong cannot be fully explained by changing emissions from biomass burning, although these likely make a substantial contribution in summer.

3.4. Anthropogenic Emissions

Thus far, we have used a variety of observational data sets to show that the observed, consistent decreasing HCHO trend cannot be explained by CH_4 oxidation, biogenic NMVOC emissions, or biomass burning emissions. We also investigated whether a change in meteorology was likely to have caused a change in the transport of HCHO or its precursors from source regions to Wollongong. We calculated trends in both wind speed and direction using BoM measurements from the nearby station at Bellambi (see Figure 1). Neither variable showed compelling evidence for a change in transport in this region. We do not have observational constraints on $^{\bullet}OH$ to test for a trend in HCHO loss, but we anticipate that any changes due to $^{\bullet}OH$ would be too small to drive the observed HCHO trend and difficult to attribute, given that an $^{\bullet}OH$ increase would be associated with both increased HCHO loss and increased HCHO production via CH_4 and NMVOC oxidation.

In the absence of other explanations, we infer that the overarching decreasing trend in HCHO must be driven by decreases in local industrial emissions of HCHO and/or its precursors. Besides the FTIR record at Wollongong, there are no long-term measurements of anthropogenic NMVOCs in this region. We instead use estimated anthropogenic emissions for 1996–2014 from the Community Emissions Data System (CEDS) inventory described by Hoesly et al. (2018) to test whether an anthropogenic emissions trend is plausible. We note that there are significant uncertainties associated with calculating local trends based on inventory estimates, especially when multiple data sources covering different time periods are used to construct the inventory. In particular, the CEDS emissions are especially uncertain for recent years, which are based on less reliable activity data and extrapolated for years beyond inventory end dates (generally 2010; Hoesly et al., 2018). Further, the CEDS estimates are scaled to Australia-specific estimates for only 3 years of the

inventory (2000, 2006, and 2012) and only at the country-wide level; downscaling from country-aggregated emissions is based on proxy data (including population and other global inventory estimates). Nonetheless, without further observational constraints, the CEDS estimates provide the best approximation of plausible changes in industrial emissions in this region.

Figure 3b shows trends in CEDS-estimated anthropogenic emissions of HCHO and select NMVOC precursors in the 0.5° by 0.5° grid square containing Wollongong; additional NMVOC precursors can be found in the supporting information. The figure shows a decline in anthropogenic emissions of all species in all months, consistent with our observation-based inference that trends in local anthropogenic emissions are responsible for the decreasing trend in HCHO.

We also used the CEDS inventory to investigate trends in estimated NO emissions and found there was no statistically significant trend at Wollongong on an annual timescale. Similarly, a recent analysis of tropospheric NO_2 columns from a self-consistent satellite data set showed no significant trend in NO_2 for nearby Sydney from April 1996 to September 2017 (Georgoulias et al., 2019). On monthly timescales, the CEDS NO emissions showed small but statistically significant increasing trends ranging from 0.4–0.6%/year (Figure S5). As HCHO production from both isoprene and other NMVOC precursors increases as NO_x increases (Wolfe et al., 2016), the stable or slightly increasing NO_x trend is inconsistent with the observed HCHO decreasing trend at Wollongong. We therefore infer that the anthropogenic NMVOC emissions are primarily responsible for the observed HCHO trend.

4. Summary and Conclusion

We have used a 20-year record of ground-based solar FTIR total column measurements from Wollongong (New South Wales, Australia) to identify a significant decreasing trend in HCHO in southeast Australia of −1.9%/year from 1996 to 2015. Monthly trends calculated from the same data set show that the HCHO trend is consistently negative throughout the year, with the exception of November when no significant trend is detected.

Using a variety of related data sets, we tested several possible explanations for the drivers of the decreasing trend at Wollongong. OMI satellite data suggested that at the larger (regional) scale, HCHO in southeast Australia is increasing rather than decreasing, likely driven by growth in CH_4 (and therefore CH_4 oxidation leading to HCHO production) in the background atmosphere. Comparison to others species available in the FTIR record (HCN, CO) and to estimated biomass burning from the Global Fire Emissions Database version 4 (GFED4s) indicated a decrease in the frequency and/or intensity of local fires that could be partially responsible for the observed HCHO changes, particularly in December. However, changes in biomass burning are insufficient to fully explain the decreasing trend, which is strengthened when periods with known fire events are excluded. In the absence of other explanations, we inferred that the overarching decreasing trend in HCHO must be driven by decreases in local industrial emissions of HCHO and/or its precursors, a hypothesis supported by consistent declines in estimated anthropogenic NMVOC emissions from the Community Emissions Data System (CEDS) inventory.

Temperature observations from across the region, including at a densely vegetated site west of Wollongong, showed a warming trend over the past 20 years in November only, coincident with the only month during which HCHO did not decrease. At this time of year, the seasonal transition from spring to summer is accompanied by increasing temperature, eventually crossing the threshold required for isoprene emission. We suggest that the observed November warming in the Wollongong region is driving an earlier onset of isoprene emissions—and with it an increase in HCHO production that is sufficient to offset the otherwise decreasing trend seen during the rest of the year. Projected future warming (Alexander & Arblaster, 2009; Hughes, 2003) could further lengthen the season during which biogenic NMVOCs are strongly emitted in this region, and the associated increase in HCHO production may eventually overwhelm the HCHO decreases that have occurred to date.

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